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(54) Polyester compositions and processes for moulding them.

(57) Compositions comprising (i) thermoplastic polyester of aliphatic diol, and dicarboxylic acid, at least 95% mole of said acid being terephthalic acid and (ii) inert particulate nucleating agent further contain, as a crystallization promoter, (iii) oligomeric polyester of organic dicarboxylic acid having 2 to 8 carbon atoms and organic polyol and having a Flory Interaction Parameter with said polyester below or equal to 0.2. The compositions are useful moulding compounds, the crystallization promoter enabling crystalline moulded products to be obtained at low mould temperatures. Moulding processes of the invention are carried out at less than 100°C mould temperatures.

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POLYESTER COMPOSITIONS AND PROCESSES FOR  
MOULDING THEM

5 This invention is concerned with polyester compositions and processes for moulding them. The field of this invention is the improvement of the crystallization rate and moulding behaviour of polyesters, in particular poly(alkylene terephthalate).

10 The use of inert particulate nucleating agents to promote crystallization of poly(alkylene terephthalate) is very well known. Mold temperatures of at least 120°C and optimumly 140°C in the case of poly(ethylene terephthalate) (PET) have generally been necessary for injection molded engineering  
15 applications. In the case of PET, because of its low rate of crystallization at desirable molding temperatures of less than 100°C, it has not attained widespread use as an engineering plastic. When PET is molded with conventional nucleating agents at temperatures below 100°C, it generally has poor  
20 physical properties, surface appearance and dimensional stability due to insufficient crystallization.

Various attempts have been made to improve the crystallization rate of PET. For example, the use of poly(ethylene oxide)  
25 or poly(propylene oxide) having hydroxyl functionality has been suggested in combination with nucleating agents; however, this method is undesirable because the PET reacts with the hydroxyl functionality to give a resin with lower molecular weight and poorer physical properties. The use  
30 of low molecular weight inert organic solvents such as benzophenone and phthalate plasticizers such as dioctyl phthalate have been suggested, but the use of such plasticizers alone

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is not desirable because they cause processing difficulties due to their high volatility under drying or processing conditions for PET, and tend to migrate to the surface of a molded specimen. Solvents such as dibutyl tin dilaurate  
5 have been suggested, but these are toxic materials and would be undesirable. U.S. Patent 3,516,957 teaches poly(alkylene terephthalates) with "a small percentage of an ester of an organic acid having from 10 to 27 carbon atoms and an alcohol having from 1 to 10 carbon atoms, e.g., methyl stearate or  
10 pentaerythritol tetrastearate, with the "cojoint presence of a synergistic agent such as an alkali metal salt or alkaline earth metal salt of an organic acid having 10 to 25 carbon atoms, e.g., sodium stearate; talc, and asbestos". The Flory interaction parameter for pentaerithritol tetra-  
15 stearate is 0.42, and for methyl stearate is 0.48.

We have now found a process and composition which can be used to enable polyesters such as PET to be molded at mold temperatures below 100°C to obtain highly crystalline parts even  
20 in thin sections the crystallization rate of the polyester being enhanced in the composition to enable this. The composition aspect of the invention can further provide polyester molding compositions which do not need to contain highly volatile solvents which tend to migrate to the surface of  
25 moldings.

The compositions of the invention comprise (i) thermoplastic polyester of aliphatic diol and dicarboxylic acid, at least 95 mole % of said acid being terephthalic acid, (ii) inert  
30 particulate nucleating agent, and (iii) oligomeric polyester as crystallization promoter, said oligomeric polyester being of organic dicarboxylic acid having 2 to 8 carbon atoms and organic polyol and having a Flory Interaction Parameter with said polyester (i) below or equal to 0.2.

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Although any linear thermoplastic polyester as described above can be used in the present invention, the preferred

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thermoplastic polyesters are PET or blends of PET with other thermoplastic polyesters. The invention is thus also applicable for example to poly(butylene terephthalate) (PBT), poly(hexylene terephthalate), poly(cyclohexylene terephthalate) and poly(cyclohexyl dimethylene terephthalate), or blends of these materials. Thermoplastic polyesters which comprise structural units derived from terephthalic acid and up to 5 mole percent of other aromatic or aliphatic dicarboxylic acids such as isophthalic, naphthalene-2,6-dicarboxylic acid or adipic acid are also suitable. The alcohol component of the thermoplastic polyester is preferably a single aliphatic diol but may also contain up to 30 mole percent of one or more other aliphatic diols. The invention is also applicable to polyesters derived from substituted acids such as hydroxy carboxylic acids in the amounts permitted above. Any two or more of these thermoplastic polyesters can be blended. For the sake of convenience hereafter these polyesters will be discussed largely in terms of PET or poly(alkylene terephthalate).

The poly(alkylene terephthalate) polyesters used as starting compounds preferably have an intrinsic viscosity of 0.4 to 1.4 grams per deciliter, and more preferably 0.6 to 0.8 grams per deciliter. For these purposes, the intrinsic viscosity is measured before extrusion in a 1% solution of phenol and tetrachloroethane, 60 : 40 at 25°C.

The inert particulate nucleating agent can be talc, kaolin, calcium carbonate, aluminum oxide, titanium dioxide, silica and/or graphite. Suitable amounts of nucleating agent to be used include .01 to 5% by weight, based on poly(alkylene terephthalate). The nucleating agent is preferably a mixture of two or more inert particulate materials of which graphite is one.

The oligomeric polyesters which have been found to be most useful in this invention have a preferred molecular weight,

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- $M_w$ , of 700 to 10,000. They must however have a Flory Interaction Parameter less than or equal to 0.2 with the poly(alkylene terephthalate), and be derived from organic dicarboxylic acid having 2 to 8 carbon atoms and organic polyol. The more preferred molecular weight is 900 to 5000, and when the acid is exclusively aliphatic, it is preferred that the oligomeric polyester have a molecular weight above 2000. Preferably the polyol is a glycol and has 2 to 15 carbon atoms. The oligomeric polyester can be terminated with either a fatty acid or an aliphatic alcohol preferably having 7 to 15 carbon atoms, or it can be un-terminated. The oligomeric polyester can also include an aromatic glycol. The hydroxyl number of the oligomeric polyester should preferably be less than 50 mg/g and more preferably less than 10 mg/g to prevent degradation of the poly(alkylene terephthalate) during melt processing or long term use at elevated temperatures. One suitable method for preparation of suitable oligomeric polyesters is disclosed by VanHook in U.S. patent 3,284,399. Several particular highly preferred oligomeric polyesters are as follows:
- Decyl terminated {ethylene adipate/phthalate (1/1)} 2.0
  - Isodecyl terminated {ethylene adipate/phthalate (1/2)} 2.7
  - Octyl/decyl (1/1) terminated {ethylene adipate/phthalate (1/2)} 2.2
  - $C_{14}$  fatty acid terminated poly (propylene adipate)
  - 2-ethyl hexyl terminated poly(butylene adipate)
  - Poly{propylene adipate/phthalate (3/1)} ,  $M_w = 1200$
  - Poly{propylene adipate/isophthalate (3/1)} ,  $M_w = 2000$
- The amount of oligomeric polyester which is generally used in the compositions is 0.1 to 10% by weight of the three components.
- Other additives may optionally be present in the compositions of the invention. Up to 50% by weight of the total composition of mineral fillers, fibrous reinforcement such as glass fibers, flame retardants, colorants, stabilizers,

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- water scavengers, processing aids, and impact modifiers can be included. The impact modifiers used are preferably among the ones disclosed in Dutch patent application 77/06327; Australian 25992/77; U.S. patent 3,864,428; and/or U.S. patent 3,919,353. Furthermore, up to 8% of an aromatic polycarbonate prepared by the reaction of phosgene with 2,2-(4,4'-dihydroxydiphenyl)-propane(bis phenol A) can be included to improve the impact strength.
- 10 Also, polyester-polyether elastomers can also be included in the composition as disclosed in our copending Application filed on the same date as this Application and claiming priority from U.S. Application 014405 filed 23 February 1979, the specification of which fully describes
- 15 the suitable polyester-polyether elastomers.

The compositions may be blended by tumbling the dry components together and melt blending by methods well known to those skilled in the art. The preferred method is extrusion

20 on a single or twin screw extruder.

It is surprising that the combination of oligomeric polyester and nucleating agent together gives such improvement to the crystallization rate of poly(alkylene terephthalate), especially PET. It is particularly surprising to find that

25 the high molecular weight oligomeric polyesters not only increase the crystallization rate of the poly(alkylene terephthalate), but they may actually increase it more than lower molecular weight organic solvents used at equal concentrations, contrary to the usual expectation for materials

30 which perform as plasticizers. Normally, low molecular weight plasticizers show better compatibility and better performance than high molecular weight plasticizers in PVC. It is also surprising that further improvements to the crystallization behavior and moldability may be obtained using

35 the combination of graphite and talc. Graphite has been named as a nucleating agent for PET by others, but it is

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surprising that it would cause a further improvement of the crystallization behavior of PET in the presence of another nucleating agent.

- 5 In this Specification and Claims appropriate ester-forming or transesterifiable derivatives of acids and alcohols are intended to be embraced by the disclosure of the acids and alcohols themselves. For example, anhydrides may often be used instead of acids in the formation of esters.

10

The following examples are presented to illustrate some embodiments of the invention, the preparation of components thereof and some comparative compositions. All parts and percentages are by weight unless otherwise indicated.

15

#### Examples

Measurements of crystallization rates, percentage crystallinity, nucleation temperatures ( $T_n$ ), and crystallization temperatures ( $T_c$ ) were all performed by differential thermal analysis. Crystallization rates were determined isothermally at 100°C on quenched samples of molten PET. The rate of crystallization is expressed as crystallization half-time,  $t_{1/2}$ , which is the time necessary for 50% of the polymer to crystallize. The degree of crystallinity of molded specimens was determined by the difference in area between melting endotherm and recrystallization exotherms according to equation 1:

$$\% \text{ Crystallinity of PET} = \frac{E \times \Delta A \times \Delta T_{\text{scale}} \times T_{\text{scale}}}{\Delta H_f(\text{PET}) \times R \times W \times W_f(\text{PET})} \times 100 \quad (1)$$

- 30 where E is an instrument correction factor,  $\Delta A$  is the difference in areas,  $\Delta T_{\text{scale}}$  is the vertical temperature response,  $T_{\text{scale}}$  is the horizontal temperature scale,  $\Delta H_f(\text{PET})$  is 27.0 cal/g., R is the heating rate, W is the sample weight and  $W_f(\text{PET})$  is the weight fraction of PET in the sample. The nucleation temperature ( $T_n$ ) was determined as the temperature where the recrystallization peak maximum occurred during 50°C/min. cooling of a molten sample. The nucleation temperature

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is a measure of the efficiency of a nucleating agent. Higher numbers indicate greater efficiency. The crystallization temperature ( $T_c$ ) was determined as the temperature where the recrystallization maximum occurred during 20°C/5 min. heating of a partly amorphous molded samples.  $T_c$  is a measure of oligomeric polyester efficiency; lower numbers indicate a greater enhancement of low temperature crystallization of the poly(alkylene terephthalate).

10 Example 1

PET (49 parts) which an intrinsic viscosity of 0.62 and 30 parts 0.48cm (3/16") chopped glass fiber strand were dried under vacuum at 120°C for 12 hours.

15 Twenty one parts of a copolymer core/shell impact modifier having a polymethyl methacrylate shell polymerized in the presence of, and grafted to, a core of polybutyl acrylate were dried under vacuum at 60°C for 12 hours. The materials were mixed together and extruded in a 2.54cm (1") single  
20 screw extruder equipped with one vacuum vent and a nitrogen blanketed feed. The extrudate was pelletized and dried again under vacuum for 6 hours at 120°C. Extruded pellets were used to obtain the rates of crystallization, nucleation temperature, and injection molding of ASTM test specimens.  
25 The material was molded on a Newbury 42.53 g (1.5-oz.) reciprocating screw injection molder with a mold temperature of 100°C and a 45-second cycle. Molded specimens were used to determine physical properties, degree of crystallinity, and crystallization temperature.

30

Example 2

A composition was prepared according to Example 1 with a mold temperature of 140°C and a 45-second cycle.

35 Example 3

A composition prepared according to Example 1 with 3% octyl/decyl (1/1 molar) terminated {ethylene adipate/phthalate (1/2)}



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2.2 as oligomeric polyester added.

Example 4

A composition prepared according to Example 1 with 0.5% talc  
5 and 3% of the oligomeric polyester of Example 3 added.

Example 5

PET, with an intrinsic viscosity of 0.62 was dried under  
vacuum at 120°C for 12 hours. The material was extruded on  
10 a 2.03cm (0.8") twin-screw extruder equipped with two vacuum  
vents and a nitrogen blanketed feed. The extrudate was  
pelletized and dried again under vacuum for 6 hours at 120°C.  
Extruded pellets were used to obtain the rates of crystall-  
ization and for injection molding of ASTM test specimens.  
15 A Newbury 42.53 g (1.5-oz.) reciprocating screw injection  
molder was used with a mold temperature of 100°C and a 45-  
second cycle. Molded specimens were used to determine phy-  
sical properties, degree of crystallinity, and crystallization  
temperature.

20

Example 6

A composition prepared according to Example 5 with 1.6% talc  
and 5.1% of oligomeric polyester octyl/decyl (1/1 molar)  
terminated {ethylene adipate/phthalate (1/2)} 2.2 added.

25

Example 7

A composition prepared according to Example 5 with 30% of the  
core/shell graft copolymer impact modifier defined in Example  
1 added.

30

Example 8

A composition prepared according to Example 5 with 28.6% of  
the core/shell graft copolymer impact modifier defined in  
Example 1, 1.1% talc, and 3.6% oligomeric polyester octyl/  
35 decyl (1/1 molar) terminated {ethylene adipate/phthalate  
(1/2)} 2.2 added.

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Examples 1 - 8 in Table I show the advantages of adding both an oligomeric polyester and a nucleating agent to PET, impact-modified PET, and impact-modified, glass-reinforced PET. The crystallization half-time decreases and percent  
5 crystallinity of the PET increases without the loss of impact strength or tensile strength when the oligomeric polyesters were added. In addition, the nucleation temperature increases when a nucleating agent was added. The lowest  
10  $t_{1/2}$  values were observed when the nucleating agent and oligomeric polyester were both added.

TABLE I

Example No.	Mold Temp. (°C)	Nucleation Temp. (°C)	t <sub>1/2</sub> (min.) at 100°C	% Crystalline 60-Mil 125-Mil Plaque Plaque	Notched Izod (ft.-lb/in)	Tensile Strength 106Nm <sup>-2</sup> (psi)
1	100	177	100	18 36	1.12 (2.1)	98.6 (14,300)
2	140	177	100	36 --	1.07 (2.0)	97.22 (14,100)
3	100	176	50	-- 38	1.07 (2.0)	96.53 (14,000)
4	100	191	16	26 35	0.91 (1.7)	95.84 (13,900)
5	100	--	43	-- 29	0.27 (0.5)	49.64 ( 7,200)
6	100	--	2.4	-- 38	0.16 (0.3)	64.81 ( 9,400)
7	100	--	88	-- 29	1.23 (2.3)	34.47 ( 5,000)
8	100	--	13	-- 37	0.8 (1.5)	29.65 ( 4,300)

5

10

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Example 9

A composition prepared according to Example 1 with 0.7% talc and 0.7% graphite added.

5 Example 10

A composition prepared according to Example 1 with 0.5% graphite and 3% oligomeric polyester oligomeric polyester octyl/decyl (1/1 molar) terminated {ethylene adipate/phthalate (1/2)} 2.2 added.

10

Example 11

A composition prepared according to Example 1 with 0.5% talc, 0.5% graphite, and 3% oligomeric polyester octyl/decyl (1/1 molar) terminated {ethylene adipate/phthalate (1/2)} 2.2 added.

15

Example 12

A composition prepared according to Example 5 with 0.8% talc, 0.8% graphite, and 5.1% of oligomeric polyester octyl/decyl (1/1 molar) terminated {ethylene adipate/phthalate (1/2)}

20 2.2 added.

Example 13

A composition prepared according to Example 5 with 28.6% the all acrylic core/shell graft copolymer impact modifier used  
25 in Example 1, 0.6% talc, 0.6% graphite, and 3.6% oligomeric polyester oligomeric polyester octyl/decyl (1/1 molar) terminated {ethylene adipate/phthalate (1/2)} added.

Examples 9 - 13 in Table II show the effect of graphite on  
30 the crystallization behavior and physical properties of PBT compositions. The effect of graphite on the crystallization half time,  $t_{1/2}$ , is particularly surprising. A comparison of Examples 4, 6, and 8 with similar compositions in Examples 11, 12, and 13, respectively, reveals that added graphite  
35 lowers the  $t_{1/2}$  in every instance.

TABLE II

Example No.	Mold Temp. (°C)	Nucleation Temp. (°C)	$t_{1/2}$ (min.) at 100°C	% Crystalline 60-Mil 125-Mil Plaque Plaque	Notched Izod Nm/cm lb/in (ft.- lb/in)	Tensile Strength $10^6 \text{ Nm}^{-2}$ (psi)
9	100	192	60	--	31	--
10	100	189	17	--	39	95.84 (13,900)
11	100	197	13	31	33	93.77 (13,600)
12	100	--	2.1	--	37	66.19 (9,600)
13	100	--	5.3	--	36	31.72 (4,600)

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Example 14

A composition prepared according to Example 1 with 5% oligo-  
meric polyester octyl/decyl (1/1 molar) terminated {ethylene  
adipate/phthalate (1/2)} 2.2, 0.75% talc, and 0.75% graphite  
5 added.

Example 15

A composition prepared according to Example 1 with 5% dibutyl  
phthalate, 0.75% talc, and 0.75% graphite added.  
10

Example 16

A composition prepared according to Example 1 with 5% dimethyl  
phthalate, 0.75% talc, and 0.75% graphite added.

15 Example 17

A composition prepared according to Example 1 with 3% oligo-  
meric polyester octyl/decyl (1/1 molar) terminated {ethylene  
adipate/phthalate (1/2)} 2.2, 0.75% talc, and 0.75% graphite  
added.

20

Example 18

A composition prepared according to Example 1 with 3%  $M_n =$   
4200 g/mol oligomeric polyester poly(propylene adipate),  
0.75% talc, and 0.75% graphite added.

25

Example 19

A composition prepared according to Example 1 with 3% Dow  
Corning 200 poly(dimethyl siloxane) silicone fluid, 0.75%  
talc, and 0.75% graphite added.

30

Example 20

A composition prepared according to Example 1 except that a  
mixture of 17.5% by weight of an MBS impact modifier con-  
sisting of a butadiene-styrene copolymer rubbery core onto  
35 which was grafted methyl methacrylate and 3.5% of bisphenol-  
A-polycarbonate was substituted for the 21% impact modifier  
defined in Example 1.

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Example 21

A composition prepared according to Example 20 with 3.5% oligomeric polyester octyl/decyl (1/1 molar) terminated {ethylene adipate/phthalate (1/2)} 2.2, 0.75% talc, and  
5 0.75% graphite added.

Example 22

A composition prepared according to Example 20 with 3.5% oligomeric polyester 2-ethylhexyl terminated poly(butylene  
10 adipate) with  $M_w = 3500$  g/mole, 0.75% talc, and 0.75% graphite added.

Example 23

A composition prepared according to Example 20 with 3.5% oligomeric polyester  $C_{16}$ - $C_{18}$  fatty acid terminated poly(propylene adipate) with  $M_w = 3400$ , 0.75% talc, and 0.75% graphite  
15 added.

Example 24

A composition prepared according to Example 20 with 3.5% epoxidized glycerol tri(linoleate), 0.75% talc, and 0.75% graphite added.  
20

Example 25

A composition prepared according to Example 20 with 3.5% of 2-ethylhexyl terminated poly(propylene sebacate) with  $M_w = 7400$ , 0.75% talc, and 0.75% graphite added.  
25

Example 26

A composition prepared according to Example 20 with 3.5% of  $C_6$ - $C_{10}$  alcohol terminated poly(butylene adipate) with  $M_w = 5000$ , 0.75% talc, and 0.75% graphite added.  
30

Example 27

A composition prepared according to Example 20 with 3.5% of  $C_{14}$  fatty acid terminated poly(propylene adipate) with  $M_w = 2200$  g/mole, 0.75% talc, and 0.75% graphite added.  
35

Example 28

A composition prepared according to Example 20 with 3.5% of C<sub>6</sub>-C<sub>10</sub> alcohol terminated poly(propylene adipate) with M<sub>w</sub> = 4200 g/mole, 0.75% talc, and 0.75% graphite added.

5  
Examples 14 - 28, in Table III, show the importance of using an oligomeric polyester with good compatibility with the PET to obtain high crystallization rates and low crystallization temperatures (T<sub>c</sub>). Compatibility is defined by the Flory  
10 interaction parameter for the interaction of oligomeric polyester and PET. The Flory interaction parameter was calculated from the Hildebrand solubility parameter for the oligomeric polyester and the PET by assuming the solubility parameter for PET to be  $\delta = 9.85$ , as shown in Moore et al, Polymer,  
15 2, 315 (1961). The following is used to calculate Flory interaction parameter:

$$X_{AB} = \frac{V_o(\delta A - \delta B)^2}{RT}$$

where X<sub>AB</sub> is the Flory interaction parameter, V<sub>o</sub> is the molar  
20 volume per repeat unit in the polymer,  $\delta A$  and  $\delta B$  are the Hildebrand solubility parameters for the oligomeric polyester and poly(alkyleneterephthalate), R is the gas constant, and T is the absolute temperature. Hildebrand solubility parameters may be calculated by the method of Small, J. Appl.  
25 Chem., 3, 71(1953). Hildebrand solubility parameters can also be found in the literature, and the values used in this specification are either literature or measured values. Low Flory values indicate increased compatibility between the oligomeric polyester and the PET. In addition to demonstrating the need for low Flory interaction parameters between  
30 oligomeric polyester and poly(ethylene terephthalate), the data in Table III shows that high values for T<sub>c</sub> are obtained when aliphatic carboxylic acids with 10 carbon atoms are used as the major carboxylic acid portion of the oligomeric  
35 polyester.



TABLE III

	Example No.	% Oligomeric Polyester Added	No. of Carbon Atoms in Carboxylic Acid Function	Flory Interaction Parameter	t <sub>1/2</sub> (min)	T <sub>c</sub> (°C)
5	14	5	8/6	0.011	8.5	N.O*
	15	5	8	0.043	12.3	129
	16	5	8	0.140	21.0	130
	17	3	8/6	0.011	11.5	123
	18	3	6	0.043	20.5	-
10	19	3	0	0.830	45.0	130
	20	0	-	-	100	136
	21	3.5	8/6	0.011	3.5	124
	22	3.5	6	0.105	-	130
	23	3.5	5	0.042	-	127
15	24	3.5	18	0.433	-	142
	25	3.5	10	0.116	-	142
	26	3.5	6	0.027	-	130
	27	3.5	6	0.11	4.8	126
	28	3.5	6	0.043	-	128

20

\*Not observed - sample was completely crystallized as molten.

Example 29

25 A composition prepared according to Example 1 with 7% decabromobiphenyl oxide and 3% antimony trioxide added.

Example 30

30 A composition prepared according to Example 1 with 7% decabromobiphenyl oxide, 3% antimony oxide, 0.5% talc, 0.5% graphite, and 3% oligomeric polyester octyl/decyl (1/1 molar) terminated {ethylene adipate/phthalate (1/2)} 2.2 were added.

35 Examples 29 and 30, in Table IV, show that rendering the composition flame resistant does not decrease the effect of the oligomeric polyester and nucleating agents on the

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crystallization half-time of the poly(ethylene terephthalate).

TABLE IV

5 Example No.	% Crystalline from Mold 60 mil Plaque	$t_{1/2}$ (min.)
29	37	100
30	44	16

10

Example 31 (comparative)

Four compositions prepared in accordance with Example 1 containing 0.5% talc and with 2% or 5% of pentaerythritol tetrastearate and 2% or 5% octyl/decyl (1/1 molar) terminated {ethylene adipate/phthalate (1/2)} 2.2 ("oligomeric") were compared, and the results reported in the following Table V. These examples show the advantage of this invention over the use of pentaerythritol tetrastearate ("PETS").

20

TABLE V

25 Additive	Amount	# of Carbon Atoms in carboxylic acid function	Flory Interaction Parameter	$t_{1/2}$ (min)	$T_{c_{OC}}$
PETS	2%	18	0.42	>100	133
PETS	5%	18	0.42	40	133
Oligomeric	2%	8/6	0.011	16.5	126
Oligomeric	5%	8/6	0.011	9.5	120

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## Claims:

1. A composition comprising (i) thermoplastic polyester of aliphatic diol and dicarboxylic acid, at least 95% mole of  
5 said acid being terephthalic acid, (ii) inert, particulate nucleating agent, and (iii) as crystallization promoter, oligomeric polyester of organic dicarboxylic acid having 2 to 8 carbon atoms and organic polyol, and having a Flory Interaction Parameter with said polyester below or equal to  
10 0.2.
2. A composition as claimed in Claim 1 wherein said polyester comprises: poly(ethylene terephthalate), poly(propylene terephthalate), poly(butylene terephthalate), poly  
15 (hexylene terephthalate), poly(cyclohexylene terephthalate), and/or poly(cyclohexyl dimethylene terephthalate).
3. A composition as claimed in any preceding Claim wherein said polyester comprises: poly(ethylene terephthalate) having  
20 an intrinsic viscosity of 0.4 to 1.4 grams/deciliter measured before extrusion in a 1% solution of phenol and tetrachloroethane 60 : 40 at 25°C.
4. A composition as claimed in Claim 3 wherein said intrinsic  
25 viscosity is 0.6 to 0.8.
5. A composition as claimed in any preceding Claim wherein said inert, particulate nucleating agent is talc, kaolin, calcium carbonate, aluminum oxide, titanium dioxide, silica,  
30 and/or graphite.
6. A composition as claimed in any preceding Claim wherein said nucleating agent is a mixture of at least two individual nucleating agents, one of which is graphite.  
35
7. A composition as claimed in any preceding Claim further

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including a core/shell impact modifier, bisphenol A polycarbonate, glass fiber reinforcement and/or flame retardant.

8. A composition as claimed in any preceding Claim wherein  
5 said oligomeric polyester is derived from dicarboxylic acid, aliphatic glycol, and has a molecular weight,  $M_n$ , of 700 to 10,000.
9. A composition as claimed in any preceding Claim wherein  
10 said oligomeric polyester is of a polyol having two hydroxyl groups and 2 to 15 carbon atoms.
10. A composition as claimed in any preceding Claim wherein  
15 said oligomeric polyester is terminated with an aliphatic alcohol or acid having 7 to 15 carbon atoms.
11. A composition as claimed in any preceding Claim wherein  
said oligomeric polyester comprises octyl/decyl (1/1) terminated {ethylene adipate/phthalate (1/2)} 2.3, 2-ethyl  
20 hexyl terminated poly(butylene adipate) and poly{propylene adipate/isophthalate (3/1)},  $M_w = 2000$ , and/or isodecyl terminated {ethylene adipate/phthalate (1/2)} 2.7.
12. A composition as claimed in Claim 8 wherein said oligomeric polyester is fatty acid terminated or alcohol terminated.  
25
13. A composition as claimed in Claim 8 derived from dicarboxylic acid, aliphatic glycol, and aromatic glycol.  
30
14. A composition as claimed in Claim 8 wherein said dicarboxylic acid is a mixture of aliphatic and aromatic dicarboxylic acids.
- 35 15. A composition as claimed in Claim 5 wherein the talc has an average particle size of less than 2 microns.

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16. A composition as claimed in Claim 5 or 6 wherein the graphite has an average particle size of less than or equal to 5 microns.

17. A composition as claimed in Claim 8, 9, 10, 12 or 13 wherein said organic dicarboxylic acid is exclusively aliphatic and the oligomeric polyester has a molecular weight of above 2000.

18. A moulding process wherein a composition as claimed in any preceding Claim is moulded at a mould temperature below 100°C.



European Patent  
Office

# EUROPEAN SEARCH REPORT

0015145

Application number

EP 80 30 0494

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl. 7)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
	<p><u>FR - A - 2 380 324</u> (BAYER)</p> <p>* Claims 1-6 *</p> <p>--</p> <p><u>DE - A - 2 348 377</u> (MONTECATINI)</p> <p>* Claims 1,5; page 5, lines 10-17 *</p> <p>----</p>	<p>1-4, 8-10</p> <p>1,5-7</p>	<p>C 08 L 67/02</p> <p>TECHNICAL FIELDS SEARCHED (Int. Cl. 7)</p> <p>C 08 L 67/02</p> <p>CATEGORY OF CITED DOCUMENTS</p> <p>X: particularly relevant A: technological background O: non-written disclosure P: intermediate disclosure T: theory or principle underlying the invention E: conflicting application D: document cited in the application L: citation for other reasons</p> <p>&amp;: member of the same patent family. corresponding document.</p>
<p>The present search report has been drawn up for all claims</p>			
Place of search	Date of completion of the search	Examiner	
The Hague	21-05-1980	DECOCKER	